POSSIBLE EXPLANATION OF ^{4}He PRODUCTION IN A Pd/D$_2$ SYSTEM BY THE TNCF MODEL

NUCLEAR REACTIONS IN SOLIDS

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Experimental data showing generation of ^{4}He from a Pd sheet-D$_2$ gas system observed by E. Botta et al. are analyzed by the trapped neutron catalyzed fusion (TNCF) model. The proposed mechanism of ^{4}He generation is not the direct d-d reaction but the reactions between the trapped neutron and a Pd isotope, n-$^{A}_{46}$Pd reactions, with a supplemental assumption, decrease of threshold energies for (n,α) reactions of $^{A}_{46}$Pd in solids. The arbitrary parameter n_n, the density of the trapped neutron, of the model is determined to be $\sim 10^{12}$ cm^{-3}, which is consistent with values determined in analyses of data in various events in the cold fusion phenomenon.

I. INTRODUCTION

It is well known now that the cold fusion phenomenon (CFP), named at first after supposition of the direct *d-d* fusion reaction in solids at room temperature, includes many "events" from the excess heat generation, tritium production, neutron emission to ^{4}He generation, and various nuclear transmutations (NTs). This diversity of the events tells us the fundamental cause of the phenomenon should not be the simple *d-d* fusion reaction but more complex reactions occurring in solids as a complex system. Therefore, the name "cold fusion" should be interpreted as "nuclear reactions and accompanying events in solids including high-density hydrogen isotopes."

The trapped neutron catalyzed fusion (TNCF) model proposed by one of the present authors (H. Kozima) has its basis on several experimental facts from the absence of events in circumstances without background neutrons and their enhancement by an artificial thermal neutron source to numerical relations between amounts of products obtained in the experiments. Ratios of the numbers of events N_M for products M calculated from experimental data are far from anticipation of the *d-d* reaction but are in good qualitative and sometimes quantitative accordance with theoretical prediction based on the TNCF model.

In electrolytic experiments with the Li electrolyte, the generation of ^{4}He has been explained by a reaction between a neutron *n* and ^{6}Li consistent with the amount of the excess heat *Q* observed simultaneously. On the other hand, in a gas contact system, there have recently been reported several data of the CFP including 2.45 MeV and higher energy neutron emission[1–3] and ^{4}He detection including those[4,5] taken up in this paper. The mechanism to produce ^{4}He in this case is different from the one in the electrolytic system with $^6_3$Li and is similar to the one resulting in NTs as explained hereafter giving a unified interpretation of several events in the CFP.

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The observation of the "2.45-MeV" neutron reported by Bressani et al.,[1] Botta et al.,[2] and Botta et al.[3] was explained qualitatively by us[6] using the TNCF model as a result of the following series of reactions in a Pd/D system not depending on the direct *d-d* fusion reaction:

$$n + d = t\,(6.98\text{ keV}) + \gamma\,(6.25\text{ MeV}) \ , \tag{1}$$

$$t(\varepsilon) + d = {}^4\text{He}\,(3.5\text{ MeV}) + n\,(14.1\text{ MeV}) \ , \tag{2}$$

$$\gamma\,(6.25\text{ MeV}) + d = n + p \ , \tag{3}$$

$$n\,(14.1\text{MeV}) + d = n' + d' \ , \tag{4}$$

and

$$n\,(14.1\text{MeV}) + d = n + p + n \ . \tag{5}$$

The cross sections of the reactions (1) through (5) are 5.5×10^{-4}, 3.0×10^{-6}, 2×10^{-3}, 0.62, and 0.18 b, respectively.

The accelerated deuteron up to an energy ϵ (the maximum value is 12.5 MeV) in the reaction (elastic collision) (4) can induce direct *d-d* reactions generating 2.45-MeV neutrons as follows:

$$d\,(\varepsilon) + d = {}^3\text{He} + n\,(2.45\text{ MeV}) + \varepsilon \ , \tag{6}$$

$$= t + p \ . \tag{7}$$

Numerical calculation of an energy spectrum of neutrons generated in the foregoing series of reactions was performed and compared with the experimental results with qualitative accordance.[6]

II. EXPERIMENTAL RESULT

The Torino group in Italy, who detected 2.45-MeV and higher energy neutrons[1–3] in Ti- and Pd-D$_2$ gas systems, also performed measurements of the ^{4}He content in samples with high loading ratio D/Pd up to 0.7 (Refs. 4 and 5). A successful measurement[4] of ^{4}He was reported at the Sixth International Conference on Cold Fusion (October 1996), and the succeeding report[5] was given at the Seventh International Conference on Cold Fusion (April 1998) with addition of a little data on the sample composition; the former of these is taken up in this paper to be analyzed by the TNCF model.

In the experiment with the observed ^{4}He in the desorbed gas from the D$_2$ loaded Pd sample, the Coehn effect was used to allow the d^+ ions to move along a Pd conductor sample from the anode toward the cathode. The sample was a Pd sheet of a size $8 \times 1 \times 1 \times 10^{-2}$ cm^3 plated by gold at both ends for a length of 1.5 cm (thickness of gold ~15 μm) and clamped there by two Cu electrodes inside the cell with a volume of 166 ± 1 cm^3.

After the vacuum control up to 1×10^{-6} mbar, D$_2$ gas was introduced in the cell to a final pressure of 2.7 bar, and the definite current up to 440 A was applied through the sample for a duration up to 2 h; at its end, the gas analysis was performed by a high-resolution mass spectrometer. In a measurement at 117 h from the D$_2$ gas introduction, where the D/Pd ratio was 0.80 ± 0.02 and the mean current 330 A (440 A at maximum) with a duration 0.4 h, an amount of $(5.3 \pm 0.7) \times 10^{18}$ ^{4}He was observed. Unfortunately enough, the excess heat to be generated accompanied with this ^{4}He production was not measured in the experiment, and it has not been improved yet in the later work reported at the Seventh International Conference on Cold Fusion.[5]

The experimental result[4] on the ^{4}He generation in the Pd/D system is analyzed in Sec. III using the TNCF model.

III. THEORETICAL EXPLANATION

There are several reactions responsible for the ^{4}He generation in Pd-D$_2$ system in the natural environment (with background neutrons) as written down as follows in addition to the aforementioned ones to explain the ~2 MeV neutrons:

$$n + d = t\,(6.98\text{ keV}) + \gamma\,(6.25\text{ MeV}) \ , \tag{8}$$

$$t\,(\varepsilon) + d = {}^4_2\text{He}\,(3.5\text{ MeV}) + n\,(14.1\text{ MeV}) + \varepsilon \ , \tag{9}$$

$$n + {}^{A}_{46}\text{Pd} = {}^{A+1}_{46}\text{Pd}^* \ , \tag{10}$$

and

$${}^{A+1}_{46}\text{Pd}^* = {}^{A-3}_{44}\text{Ru} + {}^4_2\text{He} + Q \ . \tag{11}$$

The compound nucleus ${}^{A+1}_{46}$Pd* has several channels to transform including the one taken up in the earlier reaction (11) relevant to ^{4}He generation. The threshold energies of (n, α) reactions for these isotopes ${}^{A}_{46}$Pd are a few mega-electron-volts in free space[7] as shown in Table I. The discrimination used in this paper about the decay channels will be discussed in Sec. IV.

The cross section of the first reaction (8) is 5.5×10^{-4} b and is smaller than those of the third reaction (10), with $A = 102$ to 110 leading to an unstable compound nucleus ${}^{A+1}_{46}$Pd* by a factor ~10^{-4} (compare with Table I). This is the reason that the neutron with an energy of ~2 MeV generated by the successive reactions (1) and (3) with cross sections of 0.55 and 2 mb, respectively, is smaller by a factor of 10^{-6} compared with ^{4}He generated mainly by

TABLE I

Natural Abundance, Absorption Cross Section σ (for $\frac{1}{40}$ eV neutron), Threshold Energy E_{th}, and Liberation Energy Q in Eq. (11) of Stable Pd Isotopes

	Isotope					
	${}^{102}_{46}$Pd	${}^{104}_{46}$Pd	${}^{105}_{46}$Pd	${}^{106}_{46}$Pd	${}^{108}_{46}$Pd	${}^{110}_{46}$Pd
Abundance (%)	0.96	10.97	22.23	27.33	26.71	11.81
σ (b)	3.363	0.5231	20.25	0.303	8.504	0.227
E_{th} (MeV)	2.0	4.0	3.1	6.0	9.1	11.9
Q (MeV)	4.2	6.3	3.0	2.1	1.0	1.0

the reactions (11) if its threshold energy is lowered as discussed in Sec. IV.

The number of ^{4}He atoms generated by the main reactions of n-$^A_{46}$Pd ($A = 104$ to 110) induced by the trapped thermal neutron is expressed as follows if the threshold energy E_{th} is decreased down to the thermal energy in CF materials (as discussed in Sec. IV):

$$N_{\mathrm{He}} = 0.35 n_n v_n n_{\mathrm{Pd}} V \xi \sum_A \sigma_{nA} \frac{n_A}{n_{\mathrm{Pd}}} \quad , \tag{12}$$

where $0.35 n_n v_n$ is the flow density of the thermal neutron per unit area and time, n_{Pd} is the density of Pd as a whole in the reaction region with volume V, and n_A and σ_{nA} are the density of an isotope $^A_{46}$Pd and the cross section of the reaction between n and the isotope. The factor ξ expresses an order of instability of the trapped neutron in the reaction region; we take $\xi = 0.01$ for reactions that occur in volume and $\xi = 1$ for reactions in the surface layer (where the trapped neutron reacts with lattice nuclei $^A_{46}$Pd according to the recipe of the TNCF model[8–10]).

Using the foregoing values given in Table I and assuming the reactions (10) and (11) occur in the surface layer with thickness 1 μm and $\xi = 1$, we can calculate the adjustable parameter n_n as follows:

$$n_n = 6.94 \times 10^{12}\ \mathrm{cm}^{-3} \quad . \tag{13}$$

This value is consistent with values of n_n, determined by experimental data sets obtained in the CFP, but is fairly large compared with the value $n_n = 10^4$ to 10^7 cm^{-3}, determined[6] by the experimental value N_n, i.e., number of neutrons with an energy of 2.45 MeV measured in the previous works.[1–3] The value of 10^4 cm^{-3} is in the lowest range of values obtained by us and suggests peculiarity of measurement of the neutron spectrum and/or reactions in volume with assumed value of $\xi = 0.01$.

Another possibility to remedy the difference in n_n in these events is taking ξ in the surface layer larger than 1, assumed throughout our analyses, the same value to that in vacuum. This is an attractive choice for resolution of the riddle in view of recent observation of peculiar behavior of surface nuclei showing the decay time shortening and the induced fission.[10] If we take the value of ξ in the surface layer as 10^2, the value of n_n determined by the amount of ^{4}He decreases by a factor 10^{-2}, leaving n_n determined by N_n (number of neutrons with energies at $\sim$2.45 MeV) unchanged.

If the foregoing assumption about the cause of ^{4}He generation in the Pd-D$_2$ gas system is reliable, we expect generation of Ru along with ^{4}He as observed by Botta et al.[4] It is also regrettable that we have no experimental data on the excess energy that accompanies the ^{4}He generation, which could be compared with the theoretical prediction of $Q \sim 1.8 \times 10^6$ J or 1.8 MJ deduced from the amount of ^{4}He by the reactions assumed in the explanation by the TNCF model rather than the value 2.0×10^7 J written by the authors.[4] This difference of one order of magnitude for the excess heat and also the detection of Ru in the system will give us decisive evidence about the mechanism of ^{4}He generation in the Pd-D system.

In addition to the consistent value $n_n \sim 10^{12}$ cm^{-3} obtained earlier with the values determined for other experiments,[9] it is interesting to notice that the reaction of n with $^{110}_{46}$Pd with a cross section 0.227 b and $Q = 1.0$ MeV has been used for the production of Ru in industry.

IV. DISCUSSION

After the discovery of the CFP, the cause of the various events in the phenomenon attracted the strong interest of scientists as to whether it was in the frame of quantum mechanics or not. Because the anomaly of the events is extraordinary out of understanding in ordinary physics and chemistry at first sight, there are many attempts to treat it with assumptions outside the present knowledge of quantum mechanics.

In our attempt to explain the events within a unified theoretical frame in quantum mechanics, the TNCF model has been proposed, and the model has given a consistent explanation of the CFP as a whole with a single adjustable parameter n_n, in addition to several common supplementary assumptions on the nuclear reactions in materials where the phenomenon occurs.

As is well known in neutron physics,[11] there are abundant background (BG) neutrons with thermal and epithermal energies on the earth with densities of $\sim 10^{-2}$ n/s$\cdot$cm^2 each. In view of several decisive data sets showing the absence of the CFP without BG neutrons, it is natural to pursue a possibility of explaining the phenomenon by taking into account the BG neutrons as a member of the actors playing in the drama showing abnormal events difficult to explain in a conventional frame of physics. In the TNCF model, the origin of the trapped neutron with a density n_n is assumed as the BG neutron in the first stage and in the second as neutrons generated by such breeding reactions as (2), (3), and (5) are also assumed.

The recent experimental data sets on the nuclear transmutation show clearly the existence of decay time shortening and induced nuclear fission in the system with the presence of thermal neutrons.[10,12,13] The analysis given in Sec. III also shows the reality of alpha decay of the compound nucleus, $^{A+1}_{46}$Pd* in this case, induced by the trapped neutron with thermal energy in the CF materials.

From these analyses of the events in the CFP, an interesting feature of the neutron-induced nuclear reactions is deduced: There are several channels from an initial nucleus and the trapped neutrons to the final states; for instance, in the case of the n + Pd reaction, the final states are one of those generating Zn (Ref. 14) by fission, Ag (Ref. 15) by beta decay, and Ru by alpha decay,[4] and Pd by (n,2n) reaction. The branching ratios of these channels seem variable depending on experimental conditions. While we have not determined the dependence of the branching ratios, in one case, we see Zn in Pd wire up to 40% in a 1-yr experiment[14]; in another case, we see Ag in Pd (Ref. 15); and further in the aforementioned data,[4,5] we see ^{4}He in the Pd-D_2 gas system although Ru is not yet researched in the experiment.

This experimental fact showing variable branching ratios of transmutation channels of the compound nucleus (e.g., $^{A+1}_{46}$Pd*) formed in CF materials can be taken as a signal of a new nuclear reaction in solids. Our interpretation of this fact is, as shown in several previous papers,[9,10] a nuclear transmutation of nuclei interacting with neutron Bloch waves at the crystal boundary where the probability density of the neutrons have an extreme value due to the local coherence of their wave functions. There should be a possible effect of the neutron drop[16] worked out soon. The interpretation of this fact is not fixed yet and waits for more experimental and theoretical effort.

Recent experimental data showing Zn generation in a Pd-D_2 (H_2) gas system by Mo et al.[17] together with the former data[14] have been analyzed successfully on the TNCF model, and the result will be presented elsewhere.[18] This is further evidence showing the effectiveness of the TNCF model and the reality of the aforementioned anomalous nuclear effects of the neutron Bloch waves in solids.

The neutrons with energies higher than 2.45 MeV observed often in the CF experiments and confirmed by precise measurements[1–3] will be explained by similar $(n, 2n)$ reactions as the reaction (11) expected for the neutron-Pd (or Ti) system if the (n, α) reaction (11) is conceivable in the CF material.

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